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㉗ **CONTACT LENS MATERIAL AND CONTACT LENS.**

㉘ A contact lens material comprising a copolymer prepared from a monomer mixture containing as the essential ingredient a siloxane oligomer of formula (I) and/or another siloxane oligomer of formula (II); and a contact lens made from the copolymer (I), wherein k is an integer of 1 to 3, l is 0 or 1, m is an integer of 0 to 3, n is an integer of 9 to 199, Me represents CH<sub>3</sub>, and R<sub>1</sub> represents (a), wherein R<sub>2</sub> represents H or Me, (II) wherein k is an integer of 1 to 3, l is 0 or 1, m is an integer of 0 to 3, p+q is an integer of 11 to 139 provided that p ≥ 0 and q ≥ 0, Me represent CH<sub>3</sub>, Ph represents (b), and R<sub>1</sub> is as defined in formula (I).

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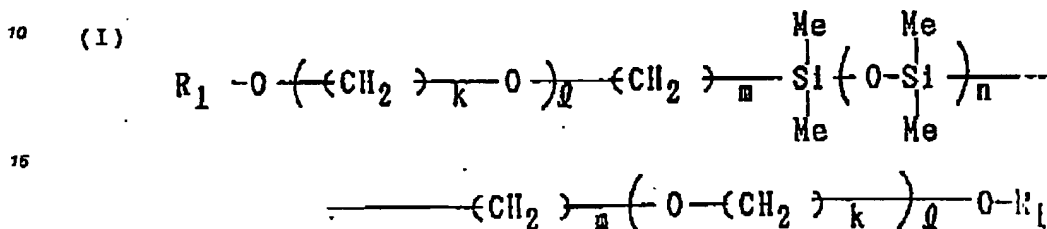




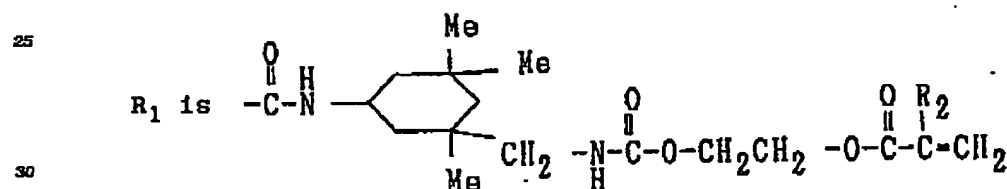
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DISCLOSURE OF THE INVENTION

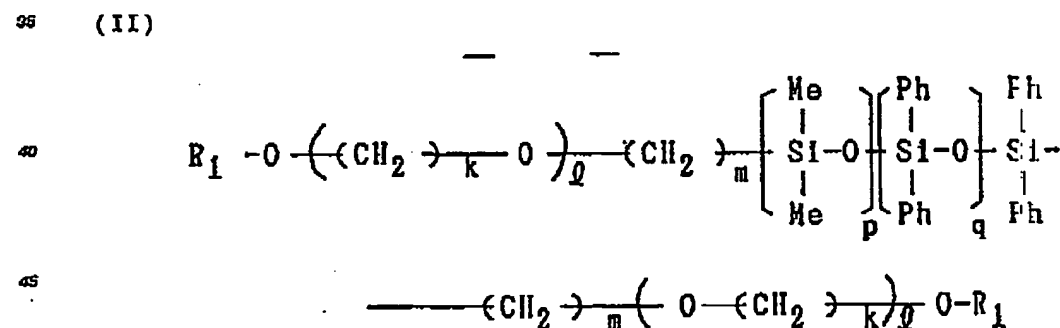
The present invention has been made in order to achieve the above object. And the present invention characteristically provides a contact lens material comprising a copolymer obtained from a monomer mixture containing, as an essential component, a siloxane oligomer of the following formula (I) and/or a siloxane oligomer of the following formula (II), and a contact lens produced by processing the above contact lens material.



[wherein k is an integer of 1 to 3, l is 0 or 1, m is an integer of 0 to 3, n is an integer of 9 to 199, Me is CH<sub>3</sub>, and



(in which R<sub>2</sub> is H or Me).



[wherein k is an integer of 1 to 3, l is 0 or 1, m is an integer of 0 to 3, p+q is an integer of 1 to 139 (provided that p≥0 and q≥0), Me is CH<sub>3</sub>, Ph is



and R<sub>1</sub> is as defined in the formula (I).]

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## MOST PREFERRED EMBODIMENTS FOR WORKING THE INVENTION

The present invention will be detailed hereinafter.

The siloxane oligomer(s) of the above formula (I) and/or the above formula (II), used in the present invention, is/are essential since these work to achieve an improvement in the shock resistance of the resultant contact lens material. Preferred are those which have a molecular weight in the range of 800 to 6,000. When the molecular weight is less than 800, the material cannot be imparted with sufficient shock resistance. When the molecular weight is more than 6,000, the material is made soft. More preferred are those which have a molecular weight in the range of 1,000 to 5,000.

The amount of the above siloxane oligomer(s) for use changes depending upon the kind, amount, etc., of monomers used for the improvement in oxygen permeability. In general, it is preferably 0.1 to 15 % by weight ("%" is used in this sense hereinafter). When this amount exceeds 15 %, the resultant polymer is undesirably liable to easily undergo plastic deformation. When it is less than 0.1 %, undesirably, no effect is produced on the improvement in shock resistance. The above amount is particularly preferably 2 to 11 %.

In the present invention, the monomer mixture for the copolymer contains at least one siloxanyl (meth)acrylate (to be referred to as "Si(M)A" hereinafter) and at least one fluorine-containing (meth)acrylate (to be referred to as "F(M)A" hereinafter) in addition to the above siloxane oligomer(s).

The Si(M)A used in the present invention is selected from trimethylsiloxydimethylsilylpropyl (meth)acrylate, bis(trimethylsiloxy)methylsilylpropyl (meth)acrylate, tris(trimethylsiloxy)silylpropyl (meth)acrylate, bis[bis(trimethylsiloxy)methylsiloxy]trimethylsiloxypropyl (meth)acrylate, bis(trimethylsiloxy)methylsiloxymonopentamethyldisiloxyimonomethylsiloxypropyl (meth)acrylate, bis(pentamethyldisiloxy)bis(trimethylsiloxy)methylsiloxypropyl (meth)acrylate, etc. In the present invention, these are used alone or in combination.

This Si(M)A is used to obtain a contact lens imparted with high oxygen permeability. The amount thereof for use is preferably 15 to 50 %. When this amount is less than 15 %, it is difficult to obtain the desired oxygen permeability. When it is more than 50 %, the copolymer is possibly softened. The amount of the Si(M)A is particularly preferably 18 to 35 %, and preferred is tris(trimethylsiloxy)silylpropyl methacrylate.

The F(M)A used in the present invention is selected, for example, from 2,2,2-trifluoroethyl (meth)acrylate, 2,2,2,2',2'',2'''-hexafluoroisopropyl (meth)acrylate, 2,2,3,3,4,4,4-heptafluorobutyl (meth)acrylate, 2,2,3,3,4,4,5,5,6,6,7,7,8,8,8-pentadecafluorooctyl (meth)acrylate, 2,2,3,3,4,4,5,5,6,6,7,7,8,8,9,9-hexadecafluorononyl (meth)acrylate, perfluorooctylethoxypropylene (meth)acrylate, perfluorooctylethoxyethylene (meth)acrylate, etc. In the present invention, these are used alone or in combination. This F(M)A not only contributes to improvement in oxygen permeability of the resultant contact lens material, but also contributes to improvement in shock resistance of the resultant contact lens material as a result of its excellent compatibility with the siloxane oligomer and the consequent promotion of dispersion of the siloxane oligomer. The amount of the F(M)A for use is preferably 15 to 60 %. When this amount is less than 15 %, the oxygen permeability is extraordinarily decreased. When it exceeds 60 %, the contact lens material is made soft. It is preferably 25 to 52 %, and the use of at least two out of the above F(M)A's is more preferred.

The monomer mixture to be copolymerized in the present invention may further contain the following monomer as required in addition to the above-specified monomers. Examples of such a monomer are alkyl (meth)acrylate (to be referred to as "R(M)A" hereinafter), a hydrophilic monomer, a crosslinking monomer, etc.

The above R(M)A may be incorporated in order to obtain a contact lens material having improved hardness. The R(M)A is selected, for example, from methyl methacrylate, ethyl methacrylate, propyl methacrylate, isopropyl methacrylate, butyl methacrylate, cyclohexyl methacrylate, 4-tert-butylcyclohexyl methacrylate, etc. These are used alone or in combination. The amount thereof for use is preferably 3 to 20 %. When this amount is less than 3 %, no effect is produced on improvement in the hardness. When it exceeds 20 %, the oxygen permeability is extraordinarily decreased. This amount is particularly preferably 4 to 16 %.

The hydrophilic monomer may be used to increase the water wettability of the contact lens material produced and improve a lens fitting sense. The hydrophilic monomer is selected, for example, from unsaturated carboxylic acids, unsaturated amides and unsaturated cyclic lactams, which are used alone or in combination. Further, in order to improve the hardness and processability of the contact lens material by improving the compatibility of the siloxane oligomer, it is preferred to use a combination of at least one unsaturated carboxylic acid with at least one unsaturated amide.

Examples of the unsaturated carboxylic acids are acrylic acid, methacrylic acid, etc., and examples of

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the unsaturated amides are acrylamide, methacrylamide, N,N-dimethylmethacrylamide, N,N-dimethylacrylamide, etc. Further, examples of the unsaturated cyclic lactams are pyrrolidone, etc. The amount of the hydrophilic monomer for use is preferably 2 to 20 %. When this amount is less than 2 %, no sufficient water wettability is obtained. When it exceeds 20 %, undesirably, not only the oxygen permeability is decreased, but also the copolymer is made soft, and further, the contact lens material is sometimes made turbid. This amount is particularly preferably 3 to 16 %.

The above crosslinking monomer is selected from tri(meth)acrylates or di(meth)acrylates of a dihydric or polyhydric alcohols, such as ethylene glycol di(meth)acrylate, diethylene glycol di(meth)acrylate, triethylene glycol di(meth)acrylate, tetraethylene glycol di(meth)acrylate, polyethylene glycol di(meth)acrylate, trimethylolpropane tri(meth)acrylate, 2-hydroxy-1,3-dimethacryloxypropane, etc. Further, allyl (meth)acrylate may be also used. These crosslinking monomers may be used alone or in combination. The crosslinking monomer is used in order to improve the contact lens material in hardness. The amount of the crosslinking monomer for use is preferably 0.2 to 15 %. When this amount is less than 0.2 %, the resultant contact lens is soft. When it exceeds 15 %, the lens material is sometimes liable to be fragile and poor in processability, or the resultant lens is sometimes liable to fracture. This amount is particularly preferably 1.5 to 8 %.

Being produced by any one of known polymerization methods, the copolymer forming the contact lens material of the present invention is particularly preferably produced by a bulk polymerization method. As a polymerization initiator for this polymerization, those which are known as a general radical generating agent can be used. The polymerization initiator is selected from peroxides such as lauroyl peroxide, cumene hydroperoxide, bis-4-tert-butylcyclohexyl peroxide, etc., and azo compounds such as azobisisobutyronitrile, azobisdimethylvaleronitrile, etc. Of these, azobisisobutyronitrile is preferred. The amount of the polymerization initiator for use is 0.05 to 0.8 % based on the total weight of the monomers.

The contact lens material of the present invention can be obtained in the state, for example, of a rod or a button by homogeneously mixing the above monomer components, casting the resultant mixture solution into a mold made of a metal, glass, plastic, etc., closing the mold, elevating the temperature in a constant-temperature bath stepwise or continuously in the temperature range between 25°C and 150°C and completing the polymerization approximately for 5 to 144 hours. In addition, it is preferred to carry out the polymerization after an inert gas such as nitrogen, argon, etc., is substituted for air in the solution and the mold is closed. The resultant copolymer as a contact lens material is finished into the form of a lens by a conventional contact lens processing method, i.e. mechanical processing procedures such as cutting, polishing, etc. In the alternative, the contact lens of the present invention may be produced by casting the monomer mixture into a mold having a predetermined radius of curvature, whereby the monomer mixture can be directly formed into a lens. In the present invention, a photopolymerization method using ultraviolet light, etc., may be also employed.

Examples of the present invention will be described below, to which the present invention shall not be limited. Physical property values and indices shown in Examples and Comparative Examples below were obtained by the following methods.

Oxygen permeability coefficient: A test piece having a thickness of 0.2 mm was measured in a 0.9 % physiological saline at 35°C with a film oxygen transmissometer of Seika type supplied by Rikaseiki Kogyo K.K.

Shock resistance test: A test piece having a diameter of 12 mm and a thickness of 2 mm was prepared and subjected to a destructive test using a Dynstat tester supplied by Toyo Seiki Seisakucho K.K. In the test, the test piece was evaluated on the basis of a shock resistance index when the energy required for the destruction of polymethyl methacrylate was taken as a shock resistance index of 100.

## [Example 1]

8.6 Percent of a siloxane oligomer [oligomer of the formula (I) wherein  $R_2 = H$ ,  $k = 3$ ,  $l = 1$ ,  $m = 3$ , and  $n = 12$ ] ("S-1" hereinafter), 19.1 % of tris(trimethylsiloxy)silylpropyl methacrylate ("Si<sub>1</sub>" hereinafter), 41.5 % of hexafluoroisopropyl methacrylate ("6F" hereinafter), 10.5 % of methyl methacrylate ("MMA" hereinafter), 11.8 % of methacrylic acid ("MA" hereinafter), 4.4 % of N,N-dimethylacrylamide ("DM" hereinafter), and 4.3 % of 2-hydroxy-1,3-dimethacryloxypropane ("HDMP" hereinafter) were mixed together. And as a polymerization initiator, 0.5 %, based on the total monomer weight, of azobisisobutyronitrile ("AIBN" hereinafter) was added to the mixture, and a homogeneous mixture was formed. Thereafter, the mixture was poured into a tube made of polyethylene, and the tube was closed. And, the temperature of the mixture was continuously elevated as follows: The tube was kept in a constant-temperature water bath at 42.5°C for 72 hours. It was further temperature-increased continuously in a hot air dryer from 42.5 to 60°C for 14 hours.

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and then from 60 to 80 °C for 10 hours. Thereafter it was kept at 80 °C for 10 hours, further temperature-increased from 80 to 100 °C for 5 hours and kept at 100 °C for 10 hours. Finally it was temperature-increased from 100 to 125 °C for 10 hours.

The resultant copolymer was colorless, transparent and optically uniform, and it also had excellent machinability in cutting, polishing, etc. The copolymer was measured for physical properties to show an oxygen permeability coefficient of  $39 \times 10^{-11}$  [mlO<sub>2</sub>(STP)/cm<sup>2</sup>·sec·mmHg] and a shock resistance index of 88. It was thus found that the copolymer was a contact lens material having good oxygen permeability and excellent shock resistance.

## 10 [Examples 2 - 33]

The procedures of Example 1 were repeated by the use of the monomer composition shown in Table 1 in amounts shown in Table 1, whereby copolymers in these Examples were obtained. Each copolymer was measured for an oxygen permeability coefficient and a shock resistance index. Table 1 shows the results.

## 15 [Comparative Examples]

The procedures of Example 1 were repeated by the use of the monomer composition shown in Table 1 in amounts shown in Table 1, whereby copolymers in these Comparative Examples were obtained. Each copolymer was measured for physical properties in the same manner as in Example 1. Table 1 shows the results.

In addition, the abbreviations used in Examples and Comparative Examples stand for the following compounds.

25	SIOL:	siloxane oligomer
	S-1:	oligomer of the formula (I) in which R <sub>2</sub> = H, k = 3, l = 1, m = 3, and n = 12.
	S-2:	oligomer of the formula (I) in which R <sub>2</sub> = H, l = 0, m = 0, and n = 12.
	S-3:	oligomer of the formula (I) in which R <sub>2</sub> = CH <sub>3</sub> , k = 3, l = 1, m = 3, and n = 13.
	S-4:	oligomer of the formula (I) in which R <sub>2</sub> = H, k = 3, l = 1, m = 3, and n = 21.
	S-5:	oligomer of the formula (I) in which R <sub>2</sub> = H, k = 3, l = 1, m = 3, and n = 55.
30	S-Z:	oligomer of the formula (III) in which R <sub>2</sub> = (b), R <sub>3</sub> = H, and t = 13.
	Si(M)A:	siloxanyl (meth)acrylate
	Si <sub>1</sub> :	tris(trimethylsiloxy)silylpropyl methacrylate
	Si <sub>2</sub> :	bis(trimethylsiloxy)methylsilylpropyl methacrylate
	Si <sub>3</sub> :	trimethylsiloxydimethylsilylpropyl methacrylate
35	Si <sub>4</sub> :	pentamethyldisiloxanylmethyl methacrylate
	F(M)A:	fluorine-containing methacrylate
	6F:	hexafluoroisopropyl methacrylate
	3F:	2,2,2-trifluoroethyl methacrylate
	FOMA:	perfluorooctylethoxypropylene methacrylate
40	FOIMA:	perfluorooctylethoxyisopropylene methacrylate
	FOEMA:	perfluorooctylethoxyethylene methacrylate
	R(M)A:	alkyl (meth)acrylate
	MMA:	methyl methacrylate
	IPMA:	isopropyl methacrylate
45	CHMA:	cyclohexyl methacrylate
	MA:	methacrylic acid
	DX:	N,N-dimethylacrylamide
	HDMP:	2-hydroxy-1,3-dimethacryloxypropane
	TMPT:	trimethylolpropane trimethacrylate
50	1G:	ethylene glycol dimethacrylate
	3G:	triethylene glycol dimethacrylate

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Table 1

MONOMER COMPOSITION (wt. %)		Examples										
		1	2	3	4	5	6	7	8	9	10	11
SIOL	S-1	8.6	8.8	8.7				8.7	11	3		3
	S-2					8.7						
	S-3						8.7					
	S-4				8.7							
	S-5										5.8	
	S-Z(*1)											
Si(M)A	Si <sub>1</sub>	19.1	19.5	20	20	20	19.7	19.7	17	25	22	30
	Si <sub>2</sub>											
	Si <sub>3</sub>											
	Si <sub>4</sub>											
F(M)A	BF	41.5	42.4	43	43	43	37	37	35	34	37.5	
	3F						13	13	10	13	7	40
	FOMA											
	FOIMA											
R(M)A	FOEMA											
	MMA	10.5	10.6	11	11	11	4.3	4.3	14.5	12	12	12
	IPMA											
Hydrophilic monomer	CHMA											
	NA	11.8	9.8	8.7	8.7	8.7	8.7	8.7	5.3	6	8.7	5
Cross-linking monomer	DX	4.4	4.5	4.3	4.3	4.3	4.3	4.3	3.7	4	4.5	5
	HDMP	4.3	4.4	4.3	4.3	4.3	4.3	4.3	3.5	3	2.5	
	TMPY											
	IG											3
	3G											
<b>PHYSICAL PROPERTIES</b>												
Oxygen permeability coefficient (*2)		39	40	38	37	40	38	37	35	44	43	42
Shock resistance index		88	85	82	84	85	87	85	84	81	83	81

\*1: conventional siloxane oligomer

\*2:  $\times 10^{-11}$  [mlO<sub>2</sub>(STP)cm/cm<sup>2</sup>·sec·mmHg]

(to be continued)



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Table 1 (continued)

		Examples												
5	MONOMER COMPOSITION (wt.%)	12	13	14	15	16	17	18	19	20	21	22		
	SIOL	S-1	5	5	9	10		8		2	8	9	8	
		S-2												
10		S-3				1								
		S-4						15						
		S-5												
		S-Z(*1)												
15	SI(N)A	SI <sub>1</sub>	25	22	25	26	25	31	15	45	30	40	31	
		SI <sub>2</sub>												
		SI <sub>3</sub>												
		SI <sub>4</sub>												
20	F(N)A	6F		38	49	50								
		3F	50											
		FOMA					23	33	50	10	30	18		
		FOIMA											50	
		FOEMA												
25	R(N)A	MMA		15		10	25	15	8	28	16	20	16	
		IPMA												
		CHMA												
30	Hydrophilic monomer	MA	10	10	12		9	8	8	9	8	4	8	
		DX	5				5	5	4	4.5	4	2	4	
	Cross-linking monomer	HDMP			5							6		
		TMPT		10		5					4		3	
		1G	5							1.5				
		3G												
35	<u>PHYSICAL PROPERTIES</u>													
	Oxygen permeability coefficient (*2)		42	51	59	52	37	40	49	35	40	38	41	
40	Shock resistance index		82	78	76	78	76	77	60	78	84	80	85	

\*1: conventional siloxane oligomer

\*2:  $\times 10^{-11}$  [mlO<sub>2</sub>(STP)cm/cm<sup>2</sup>·sec·mmHg]

(to be continued)

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Table 1 (continued)

5	MONOMER COMPOSITION (wt.%)		Examples										
			23	24	25	26	27	28	29	30	31	32	33
10	SIOL	S-1	8		8	7	3	5	8	9	8		7
		S-2											
		S-3		8								7.5	
		S-4											
		S-5											
		S-Z(*1)											
16	Si(M)A	Si <sub>1</sub>	35	30	32	40	32	32	25				23.5
		Si <sub>2</sub>								20			
		Si <sub>3</sub>									25		
		Si <sub>4</sub>										30	
20	F(M)A	8F							25		34.5		20
		3F								25		35	
		FOMA		31	25								
		FOIMA	20				30	29	10	15			
		FOEMA				20							15
26	R(M)A	MMA	20			18	22	21	17				20
		IPMA		15							15		
		CHMA			17						15	10.5	
30	Hydrophilic monomer	MA	9	9	11	10	8	8	8	9	9	9	3
		DX	5	3	3	3	2.5	2.5	3	3	4	4.5	3.5
35	Cross- linking monomer	BDMP							4				
		TMPT									4.5		3
		1G	3	3	4	2				4		3.5	
		3G					2.5	2.5					
40	PHYSICAL PROPERTIES												
	Oxygen permeability coefficient (*2)		38	49	47	39	40	43	35	33	40	34	34
45	Shock resistance index		81	80	79	79	82	81	78	80	82	85	86

\*1: conventional siloxane oligomer

\*2:  $\times 10^{-11}$  [mlO<sub>2</sub>(STP)cm/cm<sup>2</sup>·sec·mmHg]

(to be continued)

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Table 1 (continued)

MONOMER COMPOSITION (wt. %)		Comparative Examples							
		1	2	3	4	5	6	7	8
SIOL	S-1								
	S-2								
	S-3								
	S-4								
	S-5								
	S-2(*1)	5	5	5	7	10			4.8
Si(M)A	Si <sub>1</sub>	30	25	22	43	30	24	21.9	19
	Si <sub>2</sub>								
	Si <sub>3</sub>								
	Si <sub>4</sub>								
F(M)A	6F			38			56	65.8	28.6
	3F	40	50		43	45			
	FOHA								
	FOIMA								
R(M)A	FOEMA								
	MMA	12		15		10	8		28.6
	IPMA								
	CHMA								
Hydrophilic monomer	MA	5	10	10	5		8	8.8	9.5
	DX	5	5					1.75	
Cross-linking monomer	HDMP						4	1.75	
	TMPT			10		5			9.5
	1G	3	5		2				
	3G								
<b>PHYSICAL PROPERTIES</b>									
Oxygen permeability coefficient (*2)		45	42	49	55	40	60	87	21
Shock resistance index		64	62	63	60	67	55	40	70

\*1: conventional siloxane oligomer

\*2:  $\times 10^{-11}$  [mlO<sub>2</sub>(STP)/cm<sup>2</sup>·sec·mmHg]

As is clearly shown in Table 1, all the copolymers obtained in Examples 1 to 33, each of which contained the siloxane oligomer, S-1 [oligomer of the formula (I) in which R<sub>2</sub> = H, k = 3, l = 1, m = 3, and n = 12], S-2 [oligomer of the formula (I) in which R<sub>2</sub> = H, l = 0, m = 0, and n = 12], S-3 [oligomer of the formula (I) in which R<sub>2</sub> = CH<sub>3</sub>, k = 3, l = 1, m = 3, and n = 13], S-4 [oligomer of the formula (I) in which R<sub>2</sub> = H, k = 3, l = 1, m = 3, and n = 21], or S-5 [oligomer of the formula (I) in which R<sub>2</sub> = H, k = 3, l = 1, m = 3, and n = 55], exhibited a higher shock resistance index than the copolymers obtained in Comparative Examples 1 to 5, each of which contained a conventional siloxane oligomer, S-2, oligomer of the formula (III) in which R<sub>3</sub> = (b), R<sub>2</sub> = H and t = 13]. Further, the copolymers obtained in Examples 1 to 33 had an excellent oxygen permeation coefficient.

Further, in the present invention, the contact lens materials obtained in Examples, each of which contained the alkyl (meth)acrylate and/or the hydrophilic monomer and/or the crosslinking monomer exhibited a synergistic effect of these incorporated monomers and had superior advantages (e.g. hardness, water wettability, etc.). And, all the contact lens materials obtained in Examples were simultaneously excellent in many properties.

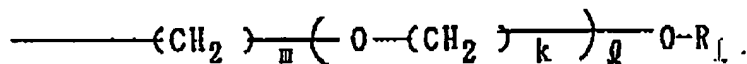
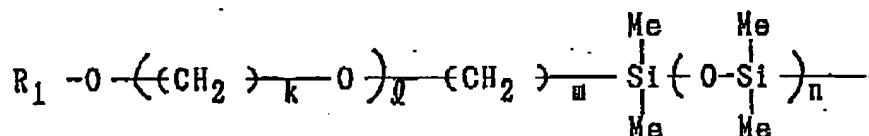
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As detailed above, due to the use of the specified siloxane oligomer, the contact lens material according to the present invention can give a contact lens which has both oxygen permeability necessary for actually fitting it on and excellent shock resistance over conventional contact lenses. Therefore, according to the present invention, the occurrence rate of destruction of lenses in handling can be decreased and the durability of lenses can be increased. As a result, the safety and economic advantages of contact lenses can be improved, and the contact lens material and the contact lens of the present invention is very useful in practical use.

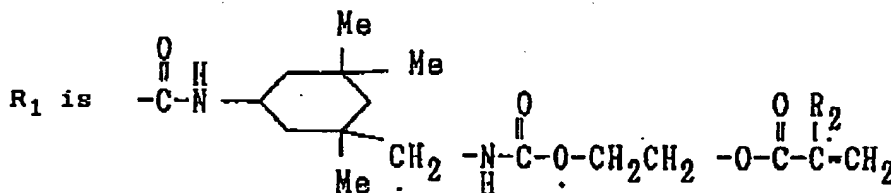
## Claims

1. A contact lens material comprising a copolymer obtained from a monomer mixture containing, as an essential Component, a siloxane oligomer of the following formula (I) and/or a siloxane oligomer of the following formula (II).

(I)

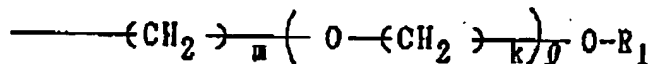
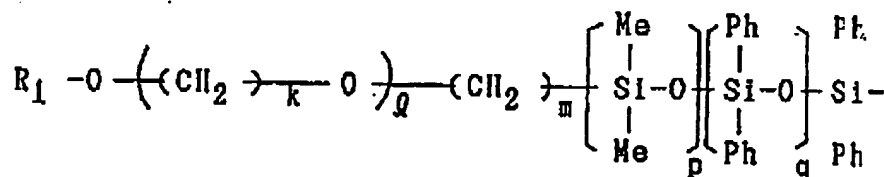


[wherein k is an integer of 1 to 3, l is 0 or 1, m is an integer of 0 to 3, n is an integer of 9 to 199, Me is CH<sub>3</sub>, and



(in which R<sub>2</sub> is H or Me)].

(II)



[wherein k is an integer of 1 to 3, l is 0 or 1, m is an integer of 0 to 3, p+q is an integer of 11 to 139 (provided that p≥0 and q≥0), Me is CH<sub>3</sub>, Ph is

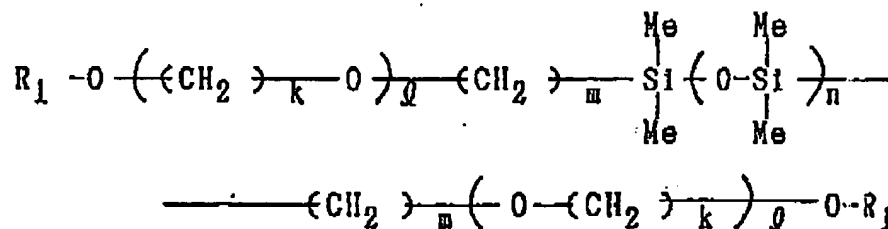
EP 0 461 270 A1



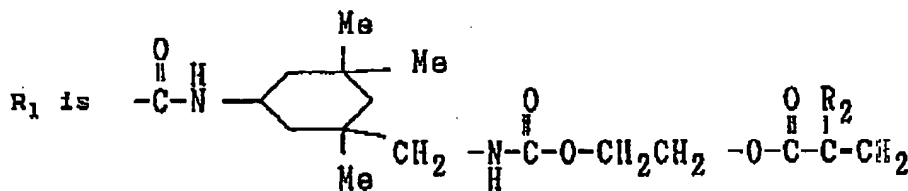
5 and  $R_1$  is as defined in the formula (I).

2. A contact lens material according to claim 1, wherein the siloxane oligomer of the formula (I) or the formula (II) has a molecular weight of 800 to 6,000.
- 10 3. A contact lens material according to claim 1, wherein the monomer mixture for the copolymer contains 0.1 to 15 % by weight of the siloxane oligomer of the formula (I) and/or the siloxane oligomer of the formula (II).
- 15 4. A contact lens material according to claim 1, wherein the monomer mixture for the copolymer is a mixture which contains, as an essential component, at least one a fluorine-containing (meth)acrylate and at least one siloxanyl (meth)acrylate.
- 20 6. A contact lens formed by processing a copolymer obtained from a monomer mixture containing, as an essential component, a siloxane oligomer of the following formula (I) and/or a siloxane oligomer of the following formula (II).

(I)



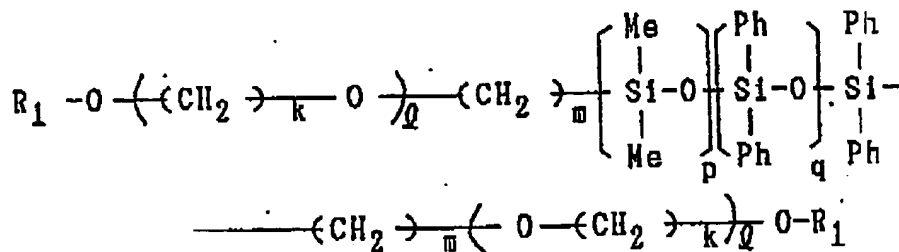
35 [wherein  $k$  is an integer of 1 to 3,  $l$  is 0 or 1,  $m$  is an integer of 0 to 3,  $n$  is an integer of 9 to 199,  $\text{Me}$  is  $\text{CH}_3$ , and



45 (in which  $R_2$  is H or Me).

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(II)



[wherein k is an integer of 1 to 3, l is 0 or 1, m is an integer of 0 to 3, p+q is an integer of 11 to 139 (provided that p≥0 and q≥0), Me is CH<sub>3</sub>, Ph is



and R<sub>1</sub> is as defined in the formula (I)].

6. A contact lens according to claim 5, wherein the siloxane oligomer of the formula (I) or the formula (II) has a molecular weight of 600 to 6,000.
7. A contact lens according to claim 5, wherein the monomer mixture for the copolymer contains 0.1 to 15 % by weight of the siloxane oligomer of the formula (I) and/or the siloxane oligomer of the formula (II).
8. A contact lens according to claim 5, wherein the monomer mixture for the copolymer is a mixture which contains, as an essential component, at least one a fluorine-containing (meth)acrylate and at least one siloxanyl (meth)acrylate.

## INTERNATIONAL SEARCH REPORT

International Application No PCT/JP90/01736

<b>I. CLASSIFICATION OF SUBJECT MATTER</b> (If several classification symbols apply, indicate all) *		
According to International Patent Classification (IPC) or to both National Classification and IPC		
Int. Cl <sup>5</sup> G02C7/04		
<b>II. FIELDS SEARCHED</b>		
Minimum Documentation Searched *		
Classification System	Classification Symbols	
IPC	G02C7/04	
Documentation Searched other than Minimum Documentation to the extent that such Documents are included in the Fields Searched *		
<b>III. DOCUMENTS CONSIDERED TO BE RELEVANT *</b>		
Category *	Citation of Document, * with indication, where appropriate, of the relevant passages **	Relevant to Claim No. **
A	JP, A, 61-138613 (Toyo Contact Lens K.K.), June 26, 1986 (26. 06. 86), Lines 4 to 19, upper part, right column, page 6 & US, A, 4,649,184 & EP, A2, 184800	1, 5
<p>* Special categories of cited documents: **</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claimed or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principles or theory underlying the invention</p> <p>"X" document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art</p> <p>"A" document member of the same patent family</p>		
<b>IV. CERTIFICATION</b>		
Date of the Actual Completion of the International Search		Date of Mailing of this International Search Report
April 1, 1991 (01. 04. 91)		April 22, 1991 (22. 04. 91)
International Searching Authority		Signature of Authorized Officer
Japanese Patent Office		

Form PCT/ISA/210 (second sheet) (January 1985)

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